Chapter 4   Appendix 1

REASSESSMENT OF GAMMA-RAY DOSES USING THERMOLUMINESCENCE MEASUREMENTS

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The epidemiological studies of atomic bomb survivors in Hiroshima and Nagasaki have provided important data about the biological effects of ionizing radiations in humans.[1-3] For these studies, the best possible estimates of radiation dose received by the organs or tissues of the survivors are required. The current individual doses of the survivors have been determined on the basis of the tentative 1965 dose (T65D).[4]

In 1976 Preeg[5] of Los Alamos National Laboratory (LANL) reported a significantly lower neutron dose in Hiroshima than the T65D estimates by calculation of neutron penetration in an infinite air medium using radiation spectra determined by a one-dimensional calculation technique. In 1978, Rossi and Mays[6] predicted a high leukemia risk for low neutron doses in occupational exposures from the results of the epidemiological study of A-bomb survivors using the T65D estimates. The two predictions posed the question whether the high leukemia risk for neutrons was valid or whether it was the result of a bias in the current A-bomb doses used by Rossi and Mays. Kerr and Pace[7] from Oak Ridge National Laboratory (ORNL) and Loewe and Mendelsohn[8] from Lawrence Livermore National Laboratory (LLNL) recalculated the gamma-ray and neutron doses in Hiroshima and Nagasaki using the spectra calculated by Preeg. The dose reassessment was discussed at the Late Effects Workshop on Dosimetry of the Atomic Bomb Survivors[9] held in conjunction with the 29th annual meeting of the Radiation Research Society in Minneapolis on 31 May 1981. At this workshop, the dosimetric techniques of T65D were reported by Auxier[4] and Maruyama.[10]

In Japan, the studies of reevaluation were started with financial help from the Ministry of Health and Welfare at the end of August 1981. The previous A-bomb dosimetries[11,12] were reexamined and their dose estimates compared with the preliminary results from LLNL and ORNL. A reassessment of gamma-ray doses was commenced using thermoluminescence (TL) measurements of ceramic materials such as bricks and tiles.

The previous gamma-ray dosimetries were carried out mostly for the survivors exposed at a distance of less than 1000 m from the hypocenter in Hiroshima and Nagasaki, because the TL apparatus previously used could not measure small TL signals. The present TL measurements were performed using recently developed apparatus and techniques. The present paper will describe the methodology of TL measurement and some dosimetric parameters required for A-bomb dosimetry; it will also present the resultant gamma-ray dose as a function of distance from the hypocenter in Hiroshima and Nagasaki.

Materials and Methods

The sampling of ceramic materials is of primary importance for the present gamma-ray dosimetry. During the past four decades, Hiroshima and Nagasaki have been amazingly reconstructed. Few buildings remain as they were at the time of the bombs. Consequently, it was not easy to collect many samples at the present time. Fortunately, a number of tile samples were collected from two Hiroshima University buildings and some brick samples were collected from a fence of a private house at Ieno-cho in Nagasaki. These buildings were located at a distance of about 1400 m from the hypocenter in both cities. During the time the present dose reassessment was being carried out, two buildings located at 500 m and 700 m from the hypocenter in Hiroshima were rebuilt and various samples were collected when the existing buildings were demolished. In Nagasaki, small pieces of brick were collected, mostly from fences of cemeteries. The samples satisfied the following conditions:

1) they were directly irradiated by the A-bomb radiations without interference of any large objects to absorb or scatter radiations,
2) fires have not occurred in or around the building from which the samples were collected,
3) there have been no artificial radiation sources around the buildings,
4) the age of the building is known,
5) the incident angle of the radiations on the samples can be easily determined, and
6) the geometry of the buildings is known precisely.

Preparation of Samples for Thermoluminescence Measurements. The distribution of the kerma-in-tissue at a point in air 1 m above the ground suggested that most of the kerma came from high-energy gamma rays.[10] According to preliminary measurements the TL at a depth of 5 mm in bricks for 40-keV x rays was three times as large as the TL for ^{60}Co gamma rays.[10] This large energy dependency is assumed not to be important for the present dose evaluation, because of the few low-energy gamma rays shown above.

The brick and decorated tile samples were prepared in the laboratory by removing 5 mm of material from the inner and outer surfaces with a diamond saw. The removal of surfaces was required for eliminating contaminations such as mud and for removing the part bleached by the sun and the part where charged-particle equilibrium may not have existed. The small pieces of brick and tile were crushed gently into grains in a mortar. Passing the crushed brick through sieves of 210 and 105 μm mesh removed grains more than 210 μm and less than 105 μm. The remaining sample with a grain size between 105 and 210 μm was washed with water and acetone in an ultrasonic cleaner. The dried samples were cleaned of

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unwanted clay matrix material by a magnetic separator. Finally, the existence of quartz in the sample was confirmed with an optical microscope. For the TL measurements the sample was divided into more than three parts to be irradiated with given doses of ^{60}Co gamma rays for calibration.

Thermoluminescence Measurements.   The present TL measurements were carried out with a Multipurpose Type 2000 TL Dosimeter (Harshaw Chemical Co., Ltd.) and all TL measurements were performed with the high temperature technique or an additive pre-dose technique.

Additive Pre-dose Technique.   In the general pre-dose technique for archaeological TL dating a radioactive beta-particle source of high dose rate is used to provide a laboratory calibration dose.[13] Since, however, the present TL measurements had to be carried out in a cold area of the laboratory, the TL samples were irradiated with doses of ^{60}Co gamma rays for the calibration before thermally activating for the pre-dose technique. The TL measurements were carried out using the TL dosimetric apparatus with a glow oven capable of heating in flowing nitrogen to 500°C maximum.

Thermal activation characteristics of brick samples differ from those of decorated tiles because of the differences in temperature of the kiln firing of the two ceramic materials. Since the maximum activation temperature of the bricks was more than 500°C, which was the maximum possible temperature of the TL dosimetric apparatus, the pre-dose technique for the brick samples was modified.

Decorative Tile Samples.   TL measurements of decorative tile samples were carried out with an additive pre-dose technique as shown in Figure 1. Three sets of powdered tile sample, seven aliquots per set, were provided for the TL measurements. One set was used for the determinations of the ratio of $S(N)/S(0)$. The remaining two sets were irradiated with different given exposures of gamma rays from a ^{60}Co teletherapy unit. Usually a beta-particle dose of 1.2 rad from a small ^{90}Sr source was used as a test dose. The ^{60}Co gamma-ray exposures were measured (in roentgens) with an ionization chamber (0.6 cc, Oyo Giken Co. Ltd.) calibrated with the National Standard Dosimeter at the Electrotechnical Laboratory at Tsukuba, Japan. Powdered samples were irradiated with gamma rays under secondary electron equilibrium by using a 1 cm thick brick plate as shown in Figure 2.

Gamma-ray doses were monitored with an MSO TL detector (MgSiO, Kasei Optinic Co. Ltd.). The dose to the quartz was calculated using a conversion factor of 0.95 rad/R for the ^{60}Co gamma rays. The doses were estimated by plotting the ratios of $S(N)/S(0)$ and $S(N+\gamma)/S(0)$ versus the test dose and the additive gamma-ray doses, including the background, and extrapolating the curves using the least-squares method.

Brick Samples.   In the TL measurements of brick samples, the dose-TL response relationship was too nonlinear to determine the gamma-ray doses with the extrapolation method using the dose-TL response curve. The profiles of sensitivity versus activation temperature for the brick samples were determined using samples kept in an oven heated for various periods. Figure 3 shows a profile of TL response versus time kept in an oven heated to 500°C. The

1. Place a sample (about 5 mg) on the dish of the TL dosimetry system using a dispenser
2. Heat to 200°C, measure S(0)
3. Heat to 500°C and keep the sample at 500°C for about 10 seconds
4. Measure S(N) or S(N+γ)
5. Determine the ratio S(N)/S(0) or S(N+γ)/S(0)

Figure 1. Protocol for the additive pre-dose technique
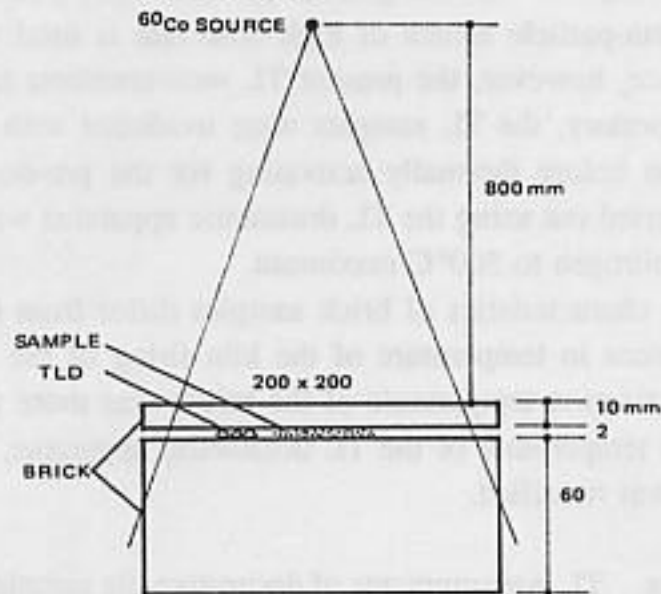

Figure 2.   Method for irradiating TL samples under radiation-equibrium conditions
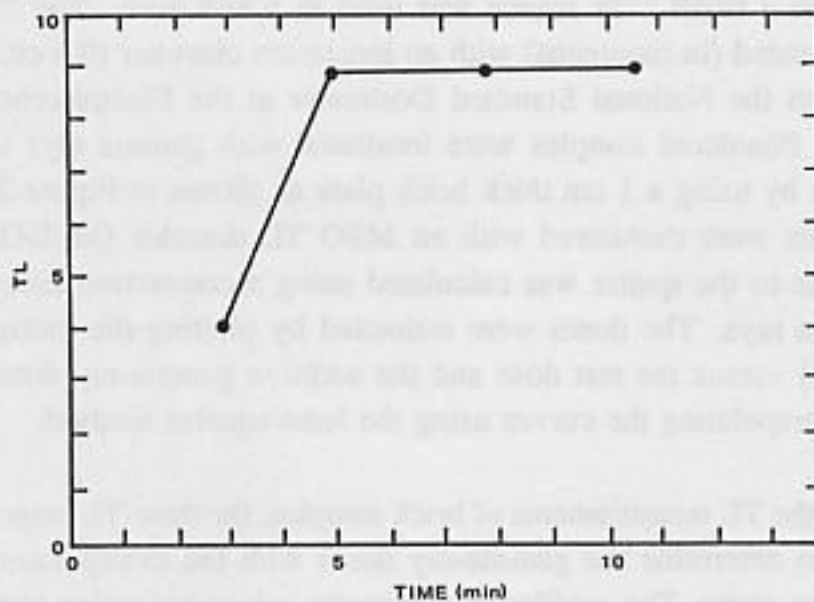

Figure 3.   Typical profile of TL response vs time kept in an oven heated to 500°C
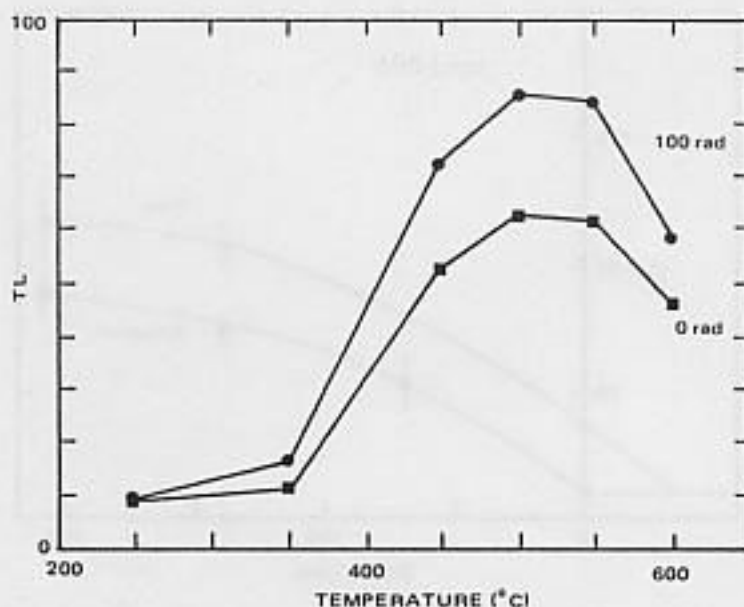
Figure 4. Profiles of TL response vs activation temperature

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1. Place sample (about 5 mg) on dish of the TL dosimetry system using a dispenser
2. Heat to 200°C, measure S(0)
3. Thermally sensitize by keeping three sets of samples in the oven heated to 525°C for 6 minutes
4. Measure S(N), S(N+γ), and S(N+2γ)
5. Anneal in oven heated to 550°C for 5 hour
6. Mix all the samples measured and divide into three sets
7. Irradiate with ⁶⁰Co gamma rays
8. Heat to 200°C, measure S'(0)
9. Thermally sensitize by keeping samples in oven heated to 525°C for 6 minutes
10. Measure S'(N), S'(N+γ), and S'(N+2γ)

Figure 5. Protocol of modified pre-dose technique for brick samples

TL response saturated when kept in the high-temperature oven for five minutes or more.

Profiles of TL response versus activation temperature were determined using brick samples without any additive dose and with 100 rad of additive dose. The results are shown in Figure 4. The TL response reached its maximum around 530°C. In this experiment, the samples were kept in various temperatures for six minutes.

The TL measurement of brick samples was carried out with a modified pre-dose technique as shown in Figure 5, taking account of the nonlinearity of TL response versus activation temperature. Three sets of powdered brick sample, seven aliquots per set, were provided for the TL measurements. One set was used for the determinations of the ratio of $S(N)/S(0)$, using the additive pre-dose technique. The remaining two sets were irradiated with different doses of gamma rays from the ⁶⁰Co unit under the same geometry shown in Figure 2. The
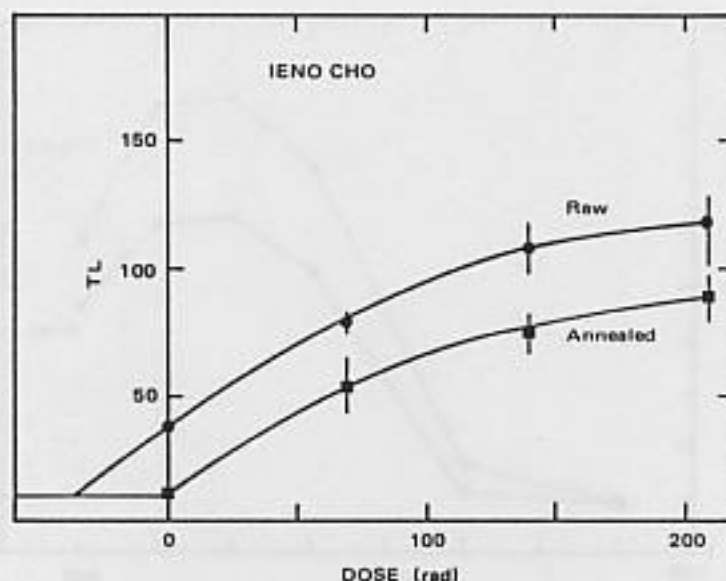
Figure 6. Example of modified additive pre-dose technique used with bricks

ratios of $S(N + \gamma)/S(0)$ and $S(N + 2\gamma)/S(0)$ were measured. After measuring the ratios, these samples were annealed at 550°C for five hours in order to completely erase associated TL signals. Annealed samples were divided into three sets and measured with the same method. These data are plotted as shown in Figure 6.

From these techniques, the gradient of the TL response versus gamma-ray dose curve around zero dose was determined. Assuming the response curve fits the function described by Chen,[14]

$$R = R_\infty \left(1 - e^{-\lambda D}\right) \tag{1}$$

where R is the TL response at dose D, R_∞ that at high dose, and λ is a constant, the parameters can be determined with the three experimental points shown in Figure 6. From this curve, it was found that the gradient of the response curves at zero dose does not vary, so this method may be useful in determining gamma-ray doses from nonlinear response curves.

High-Temperature Technique. In the pre-dose technique, linear relationships between TL response and gamma-ray dose are not obtained for the high-dose region (more than about 100 rad for brick samples and about 150 rad for the tile samples, respectively) although the dose range of the linear relationship was slightly different from sample to sample. Higher gamma-ray doses were mostly determined with a high-temperature technique using the area under the glow curves from 275 to 310°C as shown in Figure 7. In this range of temperature, the dose estimated from the area was held approximately constant with the variation of temperatue (plateau). The plateaus were confirmed in all the TL measurements with the high-temperature technique.

Background Measurements

In order to study the abundance of natural radionuclides in brick and tile samples, espe-
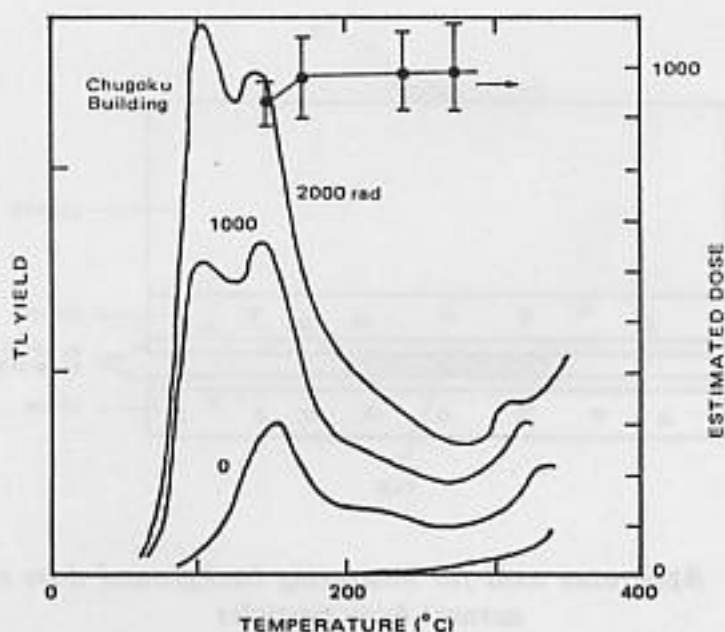
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Figure 7. Example of glow curves used in the high temperature technique

cially of alpha-particle emitters, gamma-ray spectrometry was carried out with a germanium semiconductor detector. The results suggested that brick and tile samples from Hiroshima and Nagasaki do not contain large amounts of alpha-particle emitters from the uranium and thorium series as compared with the samples used for archaeological TL dating.

The background dose from beta particles coming from natural radionuclides in brick and tile samples was measured with an experimental arrangement as shown in Figure 8. An MSO TL powder with a grain size of about 100 to 150 μm was used for the measurement of the beta-particle dose. A polyethylene foil of 100 μm thickness was used as an absorber of alpha particles from the brick and tile samples. A correction factor of 0.7 was used for the absorption of beta particles in this film. These detector assemblies were kept in a lead-shield container of 15 cm thickness for six months. The estimated background beta-ray doses are given in Table 1.

The background doses due to gamma rays from environmental radiation and from wall materials of the buildings were determined using commercially available MSO TL detectors put at various depths in an aluminum container 2 cm thick. Each dosimeter cell was left for six months on the surface of the buildings from which the samples were collected. According to measurements with an ionization chamber,[15] the annual backgrund dose from gamma rays at 1 m from the ground from environmental radiations was about 86 mrad in Hiroshima and 68 mrad in Nagasaki. The results of the TL measurements were higher than the values of environmental measurement as given in Table 1. These discrepancies may be due to natural radiations from the building materials, such as concrete, and to scattered radiations from builiding materials.

Results and Discussion

Most of the samples were decorative tiles in Hiroshima and bricks in Nagasaki. Some
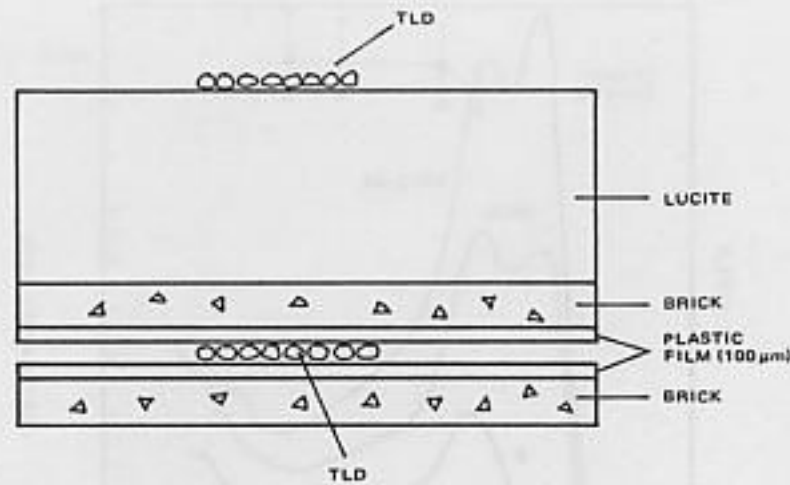
Figure 8. Apparatus used for measuring background dose rate from natural beta particles

Table 1. The Annual Natural Background Dose, Age of Building, and Tissue Dose in Air from the Atomic Bomb (Hiroshima)[a]

Place and Sample[b]	Distance (m)	Total dose (cGy)	Beta dose (Gy/a)	Gamma dose (Gy/a)	Elapsed time (a)	Bomb dose (cGy)
Naka Tel A	507	3030 ± 270[c]				3030 ± 270
B	523	3330 ± 170[c]				3330 ± 170
San'in BK.	621	1960 ± 170[c]				1960 ± 170
Chugoku E. A	665	800 ± 110[c,d]				800 ± 110
B	691	1050 ± 170[c]				1050 ± 170
H. Univ. A	1377	115 ± 7	2650 ± 320	1250 ± 40	52 ± 2	95 ± 7
B	1387	127 ± 20				107 ± 20
C	1401	11 ± 2[d]				−9 ± 3
D	1425	120 ± 13				100 ± 13
E	1426	97 ± 5				77 ± 5
F	1428	14 ± 5[d]				−6 ± 5
G	1450	123 ± 17				103 ± 17
H	1457	20 ± 4[d]				0 ± 4
R.C.Hosp.	1451	75 ± 10	2750 ± 360	1210 ± 70	44 ± 2	58 ± 10
P.S. Bureau	1613	52 ± 6	2960 ± 450	1250 ± 90	47 ± 2	32 ± 7
Prov. Dep.	3133	23 ± 5	1910 ± 380	1200 ± 80	73 ± 2	0 ± 6

[a]The dose was estimated for the samples 5-15 mm deep.
[b]The sample of the provision depot is brick, and others are ornamental tiles.
[c]High temperature peak.
[d]Shielded.

Tel.: Telephone Office; BK.: Bank; E.: Electric Company; R.C.Hosp: Red Cross Hospital; P.S.: Postal Savings; Prov. Dep.: Provisions Depot.

of these samples were of material that had been replaced after the A-bomb exposure, and their TL signals showed only the background level.

Triboluminescence resulting from the processes of crushing and mechanical handling when making powder samples was investigated using two types of brick. Some samples were irradiated with a given dose after crushing into powder and the other samples were

Table 1, Continued. (Nagasaki)[a]

Place and Sample[b]		Distance (m)	Total dose (cGy)	Beta dose (Gy/a)	Gamma dose (Gy/a)	Elapsed time (a)	Bomb dose (cGy)
Urakami		459	4430 ± 480[c]				4430 ± 480
Sakamoto		1079	775 ± 96[c]				775 ± 96
Ieno	A	1427	132 ± 21	2180 ± 280	1090 ± 30	50 ± 5	116 ± 21
	B	1427	130 ± 22				113 ± 22
	C	1437	71 ± 7[d]				55 ± 7
Zenza		1437	116 ± 24	1820 ± 220	1050 ± 60	60 ± 3	99 ± 24
Inasa	A	2026	45 ± 7	2950 ± 380	1100 ± 40	80 ± 15	13 ± 8
	B	2026	33 ± 4[d]				1 ± 5
	C	2026	27 ± 5[d]				−5 ± 6
	D	2036	32 ± 13[d]				0 ± 13
	E	2036	24 ± 8[d]				−8 ± 9
	F	2036	21 ± 7[d]				−11 ± 8
Chikugo		2369	21 ± 15	2620 ± 470	1110 ± 50	60 ± 12	−1 ± 15

[a]The dose was estimated for the samples 5-15 mm deep.

[b]The samples are brick.

[c]High temperature peak.

[d]Shielded.

Urakami: Chapel; Sakamoto: Wall around the cemetery; Ieno: Wall around the house; Zenza: Wall around the cemetery; Inasa: Ware-house; Chikugo: Wall around the cemetery

Table 2. Effects of the Sample Preparation on the High Temperature Peak (310°C)

Added dose (rad)	Irradiated after sample preparation	Irradiated before sample preparation
	Ieno-cho sample	
0	19.8 ± 1.4	
100	35.6 ± 1.7	35.2 ± 1.3
	Chugoku Electric Company sample	
0	43.6 ± 1.8	
500	79.2 ± 4.2	84.0 ± 5.4

crushed into powder after the irradiation. The TL of these samples were measured with the modified pre-dose technique and the high temperature technique. The results of the investigation are given in Table 2. It was concluded that there was no significant effect of triboluminescence in the present technique of sample preparation.

After accounting for the background dose due to beta particles and gamma rays, the gamma-ray doses from the A-bombs in Hiroshima and Nagasaki were evaluated as given in Table 1.

The annual dose from alpha particles from the clay matrix was estimated to be about a few hundred mrad per year.[16] However, in terms of the TL yield induced, the contribution by the alpha particles is much less than this value because the alpha particles are less effective in producing TL yield than are beta and gamma rays. In the pre-dose and high-temperature techniques, the TL response of alpha particles is negligibly small.
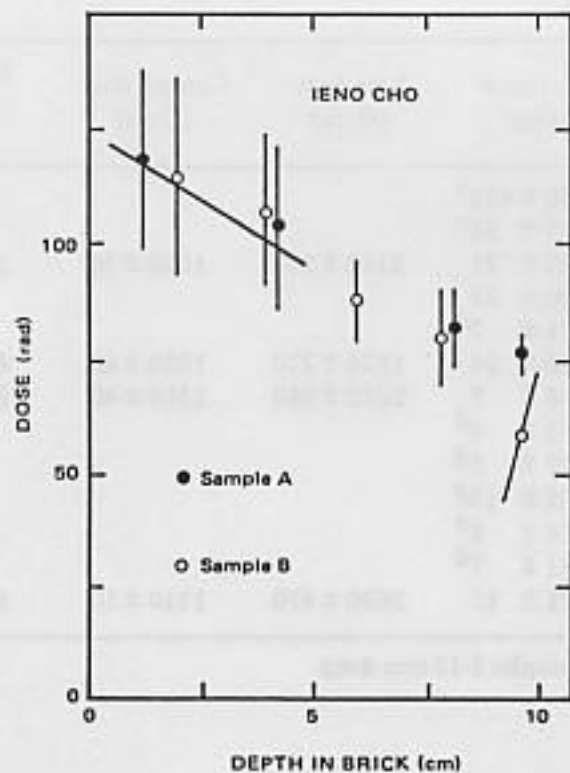
Figure 9. Depth-dose distribution in a brick from Ieno-cho. The solid
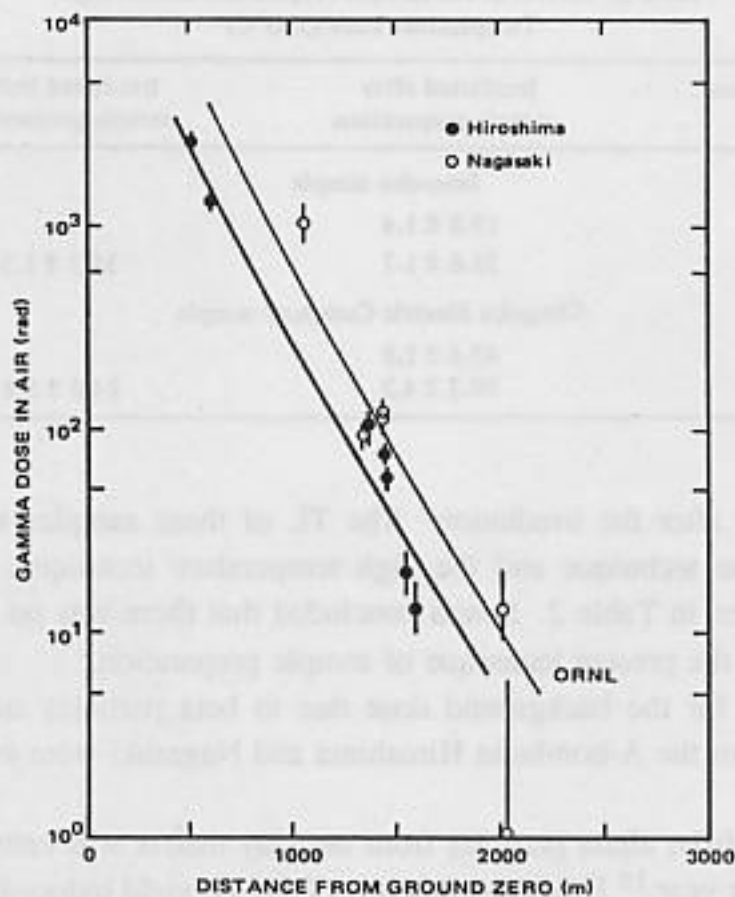line is the distribution for ^{60}Co gamma rays

Figure 10. Gamma-ray doses from the A-bombs, as determined by
thermoluminescence, as a function of distance from the hypocenter,
Hiroshima and Nagasaki

It was difficult to know precisely a time-zero point at the original firing for each brick and tile sample for the purpose of total background dose determination. Fortunately, however, knowledge of the age of the building was obtained from the register book. Before the war, the brick and decorated tile were most likely manufactured about one year before the construction of the building. In this dose estimation, the time-zero point of brick and tile samples was assumed to be one year before the age of building.

The gamma-ray doses were calculated by the following formula:

$$D = TTL - A \times TB \tag{2}$$

where D is the gamma-ray dose from the A-bomb, TTL is the total dose estimated from the TL measurement, A is the age of the brick or tile sample, and TB is the total annual dose from background beta and gamma rays.

The standard error (deviation) associated with any estimate of the A-bomb dose is then a function of the total dose estimated with the TL measurements, the age, and the annual background dose rate, and the accuracy with which each of these parameters can be measured or estimated. The standard error of the dose estimates was derived using the following formula:

$$\sigma_D = \sqrt{\sigma_{TL}^2 + \sigma_A^2 [\beta + \gamma]^2 + A^2 [\sigma_\beta^2 + \sigma_\gamma^2]} \tag{3}$$

where

σ_D = standard error of the A-bomb dose estimate
σ_{TL} = error in the total TL dose measurement
σ_A = error in age estimate for brick and tile samples
σ_β = error in annual beta-particle dose rate
σ_γ = error in annual gamma-ray dose rate.

Most of the TL measurments were carried out within standard errors less than 15%.

In order to investigate the radiation quality of the A-bomb gamma rays, the depth dose distributions in the brick sample collected from Ieno-cho in Nagasaki was measured with the additive pre-dose technique. The resultant distributions are shown in Figure 9. The solid line shows the dose distribution measured with the MSO TL detectors at various depths in a brick sample for the ^{60}Co gamma rays. From the dose distribution, it was concluded that the use of ^{60}Co gamma rays for the calibration source was appropriate for the A-bomb dosimetry.

Figure 10 shows the resultant gamma-ray doses from the A-bombs as functions of the distance from the hypocenter. The distance was determined with the army maps using the revised hypocenter in the two cities. The gamma-ray doses in Hiroshima were in good agreement with the revised doses by the ORNL group. However, in Nagasaki, the gamma-ray doses estimated by the present TL measurements agreed with the T65D estimates.

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